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Comparison of deuterium retention in tungsten pre-damaged with energetic electrons, self-ions and neutrons

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Abstract

The objective of this work is to compare the deuterium retention in tungsten pre-damaged with electron (e) beam, ions and neutrons. Self-ion irradiation was performed at IPP (Garching) and e-beam irradiation at MEPhI (Moscow). Neutron irradiation was done at Oak Ridge National Laboratory in high-flux isotope reactor (HFIR) by Hatano et al. (2013). After pre-damaging, specimens were exposed to deuterium plasma in well-defined laboratory conditions.

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Keywords: deuterium plasma; radiation-induced damage; tungsten.

1. Introduction

Due to high melting temperature, low erosion yield and low retention of hydrogen isotopes, tungsten (W) is used as plasma-facing materials in present tokamaks and selected to be used in future fusion devices, e.g. Matthews et al. (2009), Sugiyama et al. (2014), Pitts et al. (2013), or Loarer et al. (2013). Under impact of severe ITER environments, such as high heat and particle loads, neutron irradiation, etc., some important characteristics of

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tungsten can be changed significantly: retention of hydrogen isotopes, ability to reflect and absorb electromagnetic radiation emanated by plasma, erosion, surface roughness, etc. Types of radiation that can alter structural materials relevant to nuclear fusion research consist of neutrons, ions, electrons and gamma rays. All of these types of radiation have the capability to displace atoms from their lattice sites, which is the fundamental process that drives the changes in thermomechanical properties and tritium retention in structural metals. In previous works of Ogorodnikova et al. (2011), Ogorodnikova and Sugiyama (2013), O.V. Ogorodnikova and Gann (2015), Gasparyan et al. (2015), Shimada et al. (2011), Hatano et al. (2013), it was shown that pre-irradiation of W with self-ions and with neutrons at high-flux isotope reactor (HFIR) significantly increases the deuterium (D) retention in W. In the present work, we investigate the D retention in W depending on the pre-irradiation with different species.

2. Experimental

The self-ion irradiation was done at IPP (Garching) with 20 MeV W ions at different fluences ranging from 1.6×10^{16} W/m^2 to 10^{19} W/m^2, that correspond to displacement per atom (dpa) at the peak damage, from 0.005 to 3.2 dpa, respectively. The calculations were made using the methodology described in Stoller et al. (2013) and the effective threshold displacement energy of 90 eV recommended by the ASTM standard in Annual Book of ASTM Standards (1996). Irradiation with fast W ions was performed in the chamber connected to the 3 MV tandem accelerator as described in Ogorodnikova et al. (2011). The background pressure in the chamber was higher than 10^{-5} Pa. The beam was scanned over the sample surface in order to obtain a lateral homogeneous implantation. The experimental details can be found in Ogorodnikova et al. (2011, 2013, 2015). The displacement rate was ~4×10^{-3} dpa/s (approximately 0.25 dpa per hour).

An e-beam irradiation of 3.5 MeV electrons was performed at MEPhI. The irradiation was done on air with e-beam current of 40 μA/cm^2 for 3 hours. The calculations described below show that the fluence of 2.6×10^{22} e/m^2 (3 hours of irradiation) results in ~8.8×10^{-5} dpa.

Each sample was actively water cooled under irradiation and the temperature of the sample was measured by a thermocouple attached to the target holder and was kept at about room temperature under either the W ion bombardment or e-beam irradiation**.**

Neutron irradiation was done in HFIR at Oak Ridge National Laboratory (ORNL) at ambient temperature as reported in Hatano et al. (2013). Polycrystalline W targets were irradiated with mixture of fast neutrons >0.1 MeV with flux of 8.9×10^{18}m^{-2}s^{-1} and thermal neutrons with flux of 2.5×10^{19} m^{-2}s^{-1}. Due to neutron irradiation in HFIR the displacement rate is typically ~ 10^{-7} dpa/s as reported in Abromeit (1994).

After pre-damaging with either self-ions or electrons, W specimens were exposed to low-energy deuterium plasma at IPP in well-defined laboratory conditions at either 470 K or 370 K, respectively, see references of Ogorodnikova et al. (2011), (2013) and (2015). After pre-damaging with neutrons, specimens were also exposed to low-energy D plasma at Idaho National Laboratory (INL) at 470 K as reported in Hatano et al. (2013). The concentration of the retained D after the plasma exposure was examined by the nuclear reaction method (NRA) described in detail in Ogorodnikova et al. (2011), (2013) and (2015) and Hatano et al. (2013)**.**

3. Results and discussion

A comparison of the D concentration in self-ion- and n- irradiated W was made by Ogorodnikova and Gann (2015) and is presented in Fig. 1. The correlation coefficient between n- and self-ion irradiations was found by Ogorodnikova and Gann (2015) to be 0.65. This means that 1 dpa of neutrons corresponds to 0.65 ion-equivalent dpa in relation to the D retention. The conclusion was drawn in Ogorodnikova and Gann (2015) that self-ions can be used as a surrogate for a simulation of the D retention in n-irradiated W at low irradiation doses.

Electron irradiation produces damage mainly with primary knock-on energy (PKA) around the displacement threshold energy and therefore creates Frenkel defects. Therefore, e- beam with energy of 3.5 MeV produces cascade-free collisions and isolated vacancies can be created at relatively low irradiation doses.

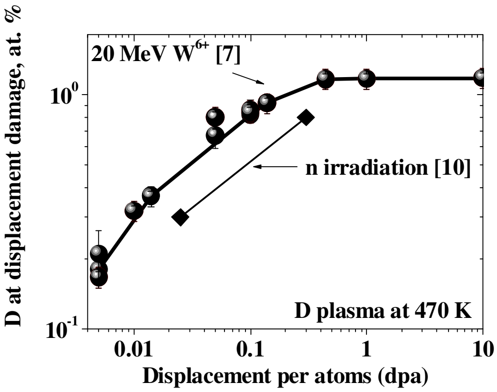

Fig. 1. Comparison of the deuterium concentration at radiation-induced defects in W created by neutron irradiation in the high-flux isotope reactor (HFIR) at Oak Ridge National Laboratory (ORNL) by Hatano et al. (2013) and by irradiation with 20 MeV W^{6+} by Ogorodnikova and Gann (2015) and subsequently exposed to D plasma at sample temperature of 470 K. Reproducible with permission from Ogorodnikova and Gann (2015)

Production of primary knock-on atom (PKA) spectrum by cascade function is known in the literature as damage energy spectrum. Damage production function for mono-energetic irradiation is expressed in Ogorodnikova and Gann (2015) by equation:

$$W(T) = \frac{1}{\sigma_D(E)} \int_{T_d}^{T} \nu(T_1) \frac{d\sigma_i(E,T_1)}{dT_1} dT_1 \tag{1}$$

where E is the ion beam energy, T is PKA energy, $d\sigma_i(E,T)/dT$ is PKA production cross-section, $\nu(T)$ is the cascade function, T_d is the threshold displacement energy for damage production.

$$\sigma_D(E) = \int_{T_d}^{T_{\max}^{(i)}} \nu(T_1) \frac{d\sigma_i(E,T_1)}{dT_1} dT_1 \tag{2}$$

is the total cross-section of defect produced by ion with an energy E, and $T_{max}^{(i)}$ is the maximal transferred energy from ion beam with an energy E (for self-ion irradiation $T_{\max}^{(i)} = E$). PKA production cross-section in the case of 20 MeV W ion irradiation can be obtained using SRIM code in Kinchin-Piese mode using effective threshold displacement energy of 90 eV. For MeV-energy range of electrons Kinchin –Piese approximation is incorrect because cascade function at low energy of PKA depends not only on energy, but also on direction of PKA in crystal lattice:

$$\nu(T,\theta,\varphi) = \Theta\left(T - T_d(\theta,\varphi)\right), \tag{3}$$

where $\Theta(x)$ is teta–function and $T_d(\theta,\varphi)$ is displacement threshold energy which depends on direction determined by polar angles θ, φ. In the paper v. Jan R et al. (1963), the following equation was suggested for $T_d(\theta,\varphi)$:

$$\begin{aligned} T_d(\theta,\varphi) = {} & (2T_{110} - T_{100}) + 2(T_{100} - T_{110})[\sin^4\theta(\cos^4\varphi + \sin^4\varphi) + \cos^4\theta] + \\ & + 9(T_{100} - 4T_{110} + 3T_{111})\sin^4\theta\cos^2\theta\sin^2\varphi\cos^2\varphi \end{aligned} \tag{4}$$

where the three parameters T_{100}, T_{110} and T_{111} represent the threshold energies along the principal crystallographic axes. For W we used T_{100}=42 eV, T_{110}=70 eV and T_{111}=44 eV reported by v. Jan et al. (1963), Singh et al. (1984), and Maury et al. (1978). Averaging $E_d(\theta,\varphi)$ over the entire solid angle of 4π, i.e. in all directions of the initial velocity of the primary knock-on atom, the crystallographic anisotropy of defect production can be eliminated and the threshold displacement energy is E_d=55 eV. Substituting (4) for (3) and averaging over all directions of PKA we obtain an average cascade function $\bar{\nu}(T)$ for polycrystalline W sample. In Fig. 2 the $\sigma_D(E)$ dependence is plotted. In the case of 3.5 MeV e-beam irradiation, the cross-section is 34 barn for the threshold displacement energy of E_d=55 eV. Therefore, 3.5 MeV e-beam irradiation up to a fluence of 2.6×10^{22} e/m^2 will create $\sigma_D(3.5)\times$fluence = 8.8×10^{-5} dpa. It should be distinguished between the effective threshold displacement energy E_d^{ef} using for cascade-full irradiation and SRIM calculations as recommended in in Stoller et al. (2013) and Annual Book of ASTM Standards (1996) and 'real' threshold displacement energy E_d. In principle, there is no any difference between effective E_d^{ef} used for SRIM calculations in the case of cascade-full irradiation and 'real' E_d used in the case of low energy e-beam irradiation generating no cascades if one uses rough sphere approximation for atom-atom potential: one has E_d for real displacement threshold and $\nu(T) = T/(2E_d)$ in formula derived by Lindhard J et al. (1963) with the same E_d for the cascade function. In fact, we have more "soft" atom-atom potential with some form-factor for cascade collisions. Norgett et al. (1975) changed Lindhard's cascade function by $\nu(T)=0.8T/(2E_d)$ and Robinson, using his cascades obtained by the molecular dynamic method, proposed to use another effective threshold displacement energy E_d^{ef} in the cascade function for large ion energies, T, and use step-function with real E_d for small T. For 3.5 MeV e-irradiation T_{max} =170 eV, it is small, neither cascades no cascade function are generated and E_d= 55 eV is more appropriate for dpa calculations.

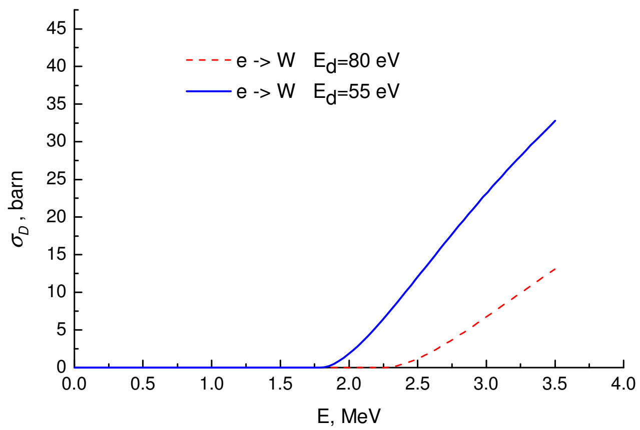

Fig. 2. Cross-section of defect production with low energy electrons using two values of the threshold displacement energy.

For fast neutrons of HFIR reactor with poly-energetic neutron flux spectrum (mixture of fast neutrons >0.1 MeV and thermal neutrons) the equation (1) must be replaced with the expression used in Ogorodnikova and Gann (2015):

$$W(T) = \frac{1}{\sigma_D}\int_0^\infty \Phi(E)\int_{T_d}^{T} \nu(T_1)\frac{d\sigma_n(E,T_1)}{dT_1}dT_1\,dE \quad . \tag{5}$$

Here Φ(E) is the differential energy spectrum of neutron flux and σ_D is the total damage production cross-section. used HFIR neutron flux spectrum given in Greenwood et al. (1985).

Fig. 3 shows the damage production functions for irradiation of W with self-ions, electrons and neutrons in HFIR. Obviously, the damage production function of self-ions reproduces better the damage function of neutrons than that of electrons. At it was pointed out in Abromeit (1994), 1 dpa electrons is equivalent to 10 neutron dpa.

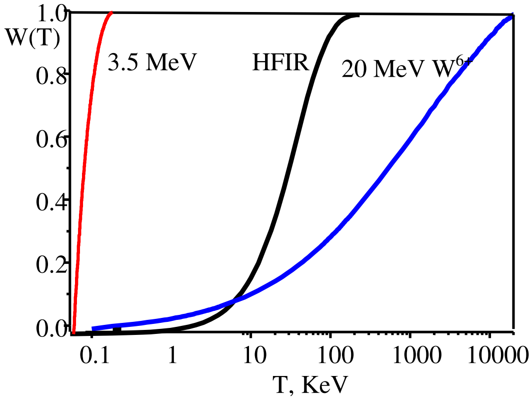

Fig. 3. Fraction of defects produced by PKA events of energies T for W irradiated with self-ions, electrons and neutrons in HFIR.

Electron irradiation creates the flat damage profile. At the same time, irradiation with 20 MeV W ions creates inhomogeneous damage profile up to ~2.5 µm with maximum at ~1.3 µm. The inhomogeneous damage profile makes the interpretation of the D depth profile data and modelling difficult. According to SRIM calculations, a four-step irradiation with W ions up to 0.45 dpa using energies of 20, 8, 4 and 2 MeV and fluences of 1.4×10^{18}, 3.06×10^{17}, 1.97×10^{17} and 1.38×10^{17} W/m^2, respectively, produces roughly rectangular damage profile as it was reported in Ogorodnikova and Gann (2015). Fig. 4 shows the D depth profile in self-ion (0.45 dpa) and e-irradiated (8.8×10^{-5} dpa) W specimens after the plasma exposure at 370 K. No remarkable increase in the D concentration in W was found in electron pre-irradiated W at dose of 8.8×10^{-5} dpa. An increase of the D concentration by two orders of magnitude in damaged zone of self-ion pre-irradiated W up to 0.45 dpa compared to undamaged W was observed that is in a good agreement with data reported in Ogorodnikova and Sugyama (2013).

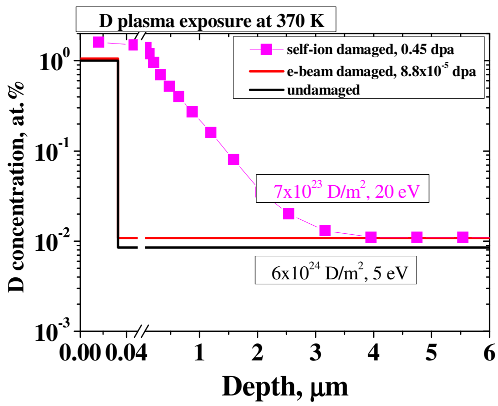

Fig. 4. A comparison of post-mortem depth profiles of deuterium in self-ion and electron pre-implanted W after exposure to deuterium plasma at 370 K. Electron irradiation was done at nearly room temperature with 3.5 MeV e^- up to a dose of 2.6×10^{22} e/m^2. Four step irradiation using four ion energies of 20, 8, 4 and 2 MeV up to an irradiation dose of $(1.4\text{-}1.6)\times10^{18}$ W/m^2 was applied in the case of self-ion irradiation to obtain flat damage profile.

For the present experiments, the e-irradiation dose was not sufficient to produce remarkable damage in W that is in an agreement with theory. Future work is in progress to compare the D retention at radiation-induced defects produced in cascade-free and cascade-full conditions.

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